Final Report: Summary of Research

Coupling Satellite and Ground-Based Instruments to Map Climate Forcing by Anthropogenic Aerosols

by Professor Robert J. Charlson (P.I.) and Theodore L. Anderson University of Washington Seattle, WA 98195

Grant Specifications:

NASA Grant: NAG-1-1877

PI: Robert J. Charlson

Period: 12/16/96 - 8/15/99 (2 years plus 8 month no-cost extension)

Amount: \$158,469

NASA Technical Officer: Chris Hostetler

University of Washington Budget Number: 61-8393

Table of Contents

BSTRACT	
BACKGROUND AND RATIONALE	2
PROGRESS	3
Task 1 Determining lidar ratios from existing in situ optical measurements	3 3 4
REFERENCES CITED	
TABLES	8
FIGURES	

Abstract

Climate forcing by anthropogenic aerosols is a significant but highly uncertain factor in global climate change. Only satellites can offer the global coverage essential to reducing this uncertainty; however, satellite measurements must be coupled with correlative, in situ measurements both to constrain the aerosol optical properties required in satellite retrieval algorithms and to provide chemical identification of aerosol sources. This grant funded the first two years of a three-year project which seeks to develop methodologies for combining spaceborne lidar with in-situ aerosol data sets to improve estimates of direct aerosol climate forcing. Progress under this two-year grant consisted in the development and deployment of a new in-situ capability for measuring aerosol 180° backscatter and the extinction-to-backscatter ratio. This new measurement capacity allows definitive lidar/in-situ comparisons and improves our ability to interpret lidar data in terms of climatically relevant quantities such as the extinction coefficient and optical depth. Measurements were made along the coast of Washington State, in Central Illinois, over the Indian Ocean, and in the Central Pacific. Thus, this research, combined with previous measurements by others, is rapidly building toward a global data set of extinctionto-backscatter ratio for key aerosol types. Such information will be critical to interpreting lidar data from the upcoming PICASSO-CENA1, or P-C, satellite mission. Another aspect of this project is to investigate innovative ways to couple the lidar-satellite signal with targeted in-situ measurements toward a direct determination of aerosol forcing. This aspect is progressing in

¹ Pathfinder Instruments for Cloud and Aerosol Spaceborne Observations - Climatologie Etendue des Nuages et des Aerosols

collaboration with NASA Langley's P-C lidar simulator and radiative transfer modeling by the University of Lille, France.

Background and Rationale

Progress on the problem of climate forcing by anthropogenic aerosols will require global-scale observations, which, in turn, can only be provided by satellite-borne instruments. With this in mind, a joint U.S./French research satellite project, PICASSO-CENA, or P-C, is being developed for a 3-year mission beginning in 2003. Following the recommendation of the U. S. National Research Council (NRC, 1996), this project will feature Light Detection and Ranging (lidar) instrumentation for obtaining quantitative, vertically resolved aerosol information both day and night, over both continents and oceans. The capabilities of spaceborne lidar have been studied previously via deployment on the U. S. Space Shuttle (Winker et al., 1996).

Lidar has become a central technology in current strategies for tropospheric aerosol research because of its demonstrated ability to map aerosol variations throughout the atmospheric column. Its use is complicated, however, by the fact that the lidar signal contains a convolution of two basic optical properties of the aerosol particles: the 180° backscatter coefficient, β_p (m⁻¹ sr⁻¹) and the extinction coefficient, σ_{ep} (m⁻¹). A quantitative retrieval of either property requires knowledge their relationship along the laser path. A central goal of this project is to review and extend current knowledge of this relationship, which we express in the usual form of an extinction-to-backscatter ratio, $S = \sigma_{ep} / \beta_p$, with units of sr. (To make this definition unambiguous, the value of S in the Rayleigh limit is $8\pi/3$.)

Like other aerosol optical properties, S can be calculated from Mie Theory. Mie calculations are essential for instrument validation experiments (e.g. Fig. 1) and provide guidance to field measurements by suggesting the range of values to be expected and what the controlling factors ought to be. A study by Ackermann (1998) illustrates the latter point. He used assumptions for aerosol size distribution and refractive index to show that S should vary from about 15 to 75 sr for tropospheric aerosols, with lower values associated with weakly absorbing coarse-mode particles (i.e. seasalt and mineral dust) and higher values associated with small and/or highly absorbing accumulation-mode particles. These Mie calculations were combined with models of aerosol hydration to develop functional relationships between S and ambient relative humidity for several aerosol types.

While valuable, such calculations are inadequate in themselves for two reasons. First, Mie calculations are limited to simple, frequently unrealistic particle morphologies (homogeneous spheres, concentric spheres, and a few variants). Second, this "theoretical" approach to determining S can never be more accurate than the *empirical* knowledge of particle

size and refractive index (the latter from knowledge of particle chemistry) upon which the calculation is based. For these reasons, direct empirical determinations of S are required.

Several methods have been developed to make this measurement; however, as shown in Table 1, below, they have been deployed to date in only a handful of investigations of tropospheric aerosols. Most regions of the Earth and many major aerosol types have no published data at all and a statistically significant database exists for only one location - Tucson, Arizona. This project seeks to expand that data base in preparation for the upcoming PICASSO-CENA satellite mission. The last four entries in Table 1 show the progress that has been made so far.

Progress

This project was originally conceived in terms of three tasks. Progress is therefore grouped under those original headings, although the direction of research has clearly changed in light of the initial findings.

Task 1 Determining lidar ratios from existing in situ optical measurements

This task was superceded by the progress under Task 2. Results from direct, calibrated measurements showed that existing, nephelometric data sets combined with Mie Theory do not offer an accurate means of predicting the lidar ratio. Therefore, our focus has shifted to expanding the data base of direct measurements.

Task 2 Design of a new instrument for directly measuring 180-backscatter

Under this task we designed, built, calibrated, and deployed a new, nephelometer-type instrument for performing direct, calibrated measurements of 180° backscatter and the extinction-to-backscatter ratio, S. Results have been reported in one published article (Doherty et al., 1999) and one article submitted for publication (Anderson et al., 2000). A summary of those, plus more recent, results in included in this section.

Figure 1 shows the calibration of the device with laboratory particles of known size and refractive index (latex spheres). The continuous lines represent Mie Theory prediction. Measured points are all seen to fall within experimental uncertainties, indicated by horizontal and vertical error bars. Based on this well-calibrated methodology, measurements were made at four sites in different regions of the globe. Results are summarized in Table 1, which serves to document as well the paucity of previous measurements of S..

Task 3. Analyze 180-backscatter optical depth as an index of aerosol climate forcing

This task has evolved into a collaborative project involving the lidar simulator model developed at NASA Langley as well as radiative transfer modeling at the University of Lille, France. Progress to date has involved developing a set of atmospheric aerosol "scenes" to be jointly studied by the lidar simulator and the radiative transfer models, analyzing preliminary results from simple tests, and designing future simulation experiments. Considerable progress on this task is expected in year three of the project, which is funded under a separate grant and will thus be the subject of a future report.

A critical underpinning to these experiments is accurate knowledge of P-C noise uncertainty and how this effects the spatial resolution of aerosol backscatter data. The remainder of this section details progress made in that area.

The effective spatial resolution of P-C lidar for aerosol studies is limited by instrumental noise, which has now been estimated via the lidar simulator. Results for the 532 nm from initial runs using two simple aerosol models imply that the noise is well behaved for clear-sky conditions and at modest levels of aerosol extinction. The equations below describe a simple model which appears to fit the simulator data quite well.

Backscatter by aerosol particles, β_a , is derived as total measured backscatter, β_m , minus the calculated molecular backscatter, β_m ,

$$\beta_{a} = \beta - \beta_{m} \tag{1}$$

Therefore, the uncertainty in β_a , which I will denote $\delta \beta_a$, is,

$$\delta \beta_a = \sqrt{\delta \beta^2 + \delta \beta_m^2} \tag{2}$$

Assuming good knowledge of molecular density, the uncertainty in β_m is just due to the calibration uncertainty, δC ,

$$\delta \beta_{\rm m} = \frac{\delta C}{C} \beta_{\rm m} \tag{3}$$

We set $\delta C/C$ to 10% for this calculation. We use the simulator results for the elevated layers of "dust" and "dirty pollution" to estimate $\delta \beta$, using a criterion of one standard deviation (70% confidence) of the simulated power for various averaging scales with respect to the grand average over the entire aerosol layer (3-5 km) and over all simulated "shots". To a very good approximation, the standard deviation of the simulated power scales with the square root of the signal, such that we can use the noise uncertainty for molecular backscatter as a reference point,

$$\delta\beta = \delta\beta_{\rm m} \sqrt{\frac{\beta}{\beta_{\rm m}}} \tag{4}$$

In addition, simulated noise scales inversely with the square root of n, the number of "bins" over which the signal is averaged. Here we use the word "bin" to refer to the highest spatial resolution

of a single shot at 30 m vertical resolution. The results in the lower troposphere are very nearly fit by,

$$\frac{\delta \beta_{\rm m}}{\beta_{\rm m}} = \frac{1}{\sqrt{n}} \tag{5}$$

Combining (2) - (5) and dividing through by $\beta_{\rm a}$,

$$\frac{\delta \beta_{a}}{\beta_{a}} = \frac{\beta_{m}}{\beta_{a}} \sqrt{\frac{1}{n} \frac{\beta}{\beta_{m}} + \left(\frac{\delta C}{C}\right)^{2}}$$
 (6)

Figure 2 shows contours of $\delta \beta_a/\beta_a$ plotted as a function of n (on the x-axis) and the magnitude of β_a (on the y-axis). The y-axis is actually β_a scaled by molecular backscatter, since this is a more meaningful quantity to most people. Given knowledge of the extinction-to-backscatter ratio, S_a , the magnitude of β_a can be converted to an aerosol extinction value, σ_a ,

$$\sigma_a (km^{-1}) = 0.00163 \frac{\beta_a}{\beta_m} S_a$$
 (6)

(Here, the aerosol extinction is implicitly referenced to standard temperature and pressure, since $0.00163 \text{ km}^{-1} \text{ sr}^{-1}$ is the 532 nm molecular backscatter at STP.) The right-hand y-axis shows the aerosol extinction level for $S_a=60 \text{ sr}^{-1}$, a typical value for pollution aerosol. For this value of S_a , $\beta_a/\beta_m=1$ corresponds to an extinction level of 0.1 km^{-1} , a very high level of extinction. The green box on Figure 2 indicates the region where reasonable accuracy (5-20%) is achieved for reasonable aerosol extinction levels (0.01 to 0.1 km⁻¹). According to these (*very preliminary*) calculations, averaging over 100 to 1000 "bins" will be necessary for accurate measurements of pollution aerosol backscatter outside of intense plumes. To put this in context, 200-bin averaging would correspond to averaging over 150 m in the vertical (5-vertical bits) and 10 km in the horizontal (40-shots).

Based on this knowledge of the effective spatial resolution of the P-C aerosol backscatter data, we are in the process of designing a correlative in-situ measurement strategy for the P-C mission. We expect to submit this strategy for publication - and thus for consideration by the worldwide aerosol measurement community - within the next few months.

Refereed publications

Research efforts supported by this grant are reported in the following articles, published or submitted for publication to refereed journals. Members of the Charlson research group are indicated in bold type.

Anderson, T. L. and Ogren, J. A. (1998), Determining aerosol radiative properties using the TSI 3563 integrating nephelometer, <u>Aerosol Sci. Technol.</u> 29, 57-69.

Anderson, T. L., Covert, D. S., Wheeler, J. D., Harris, J. M., Perry, K. D., Trost, B. E. and Jaffe, D. J. (1999), Aerosol backscatter fraction and single scattering albedo: measured

- values and uncertainties at a coastal station in the Pacific Northwest, <u>J. Geophys. Res.</u>, 104, 26793-26807.
- Anderson, T. L., Masonis, S. J., Covert, D. S., Charlson, R. J. and Rood, M. J., 2000: Insitu measurement of the aerosol extinction-to-backscatter ratio at a polluted, continental site, J. Geophys. Res., submitted, April, 2000.
- Bond, T. C., Charlson, R. J. and Heintzenberg, J. (1998), Quantifying the emission of light-absorbing particles: Measurements tailored to climate studies, Geophys. Res. Lett. 25, 337-340.
- Bond, T. C., Anderson, T. L., and Campbell, D. (1999), Filter-based measurement of light absorption by particles: intercomparison and uncertainties, <u>Aerosol Sci. Technol.</u>, 30, 582-600.
- Boucher, O., Schwartz, S. E., Ackerman, T. P., Anderson, T. L., Bergstrom, B., Bonnel, B., Chylek, P., Dahlback, A., Fouquart, Y., Fu, Q., Halthore, R. N., Haywood, J. M., Iversen, T., Kato, S., Kinne, S., Kirkevåg, A., Knapp, K. R., Lacis, A., Laszlo, I., Mishchenko, M. I., Nemesure, S., Ramaswamy, V., Roberts, D. L., Russell, P., Schlesinger, M. E., Stephens, G. L., Wagener, R., Wang, M., Wong, J. and Yang, F. (1998), Intercomparison of models representing shortwave radiative forcing by sulfate aerosols, J. Geophys. Res. 103, 16979-16998.
- Charlson, R. J., Anderson, T. L. and Rodhe, H. (1999), Direct climate forcing by anthropogenic aerosols: quantifying the link between radiation and sulfate models, <u>Beitr.</u> Phys. Atmosph., 72, 79-94.
- Doherty, S., Anderson, T. L. and Charlson, R. J. (1999), Measurement of the lidar ratio for atmospheric aerosols using a 180°-backscatter nephelometer, <u>Appl. Optics</u>, 38, 1823-1832.
- Heintzenberg, J., Charlson, R. J., Clarke, A. D., Liousse, C., Ramaswamy, V., Shine, K. P., Wendisch, M. and Helas, G. (1997), Measurement and modelling of aerosol single-scattering albedo: Progress, problems and prospects, <u>Beitr. Phys. Atmosph.</u> 70, 249-263.

References Cited

- Ackermann, J., 1998: The extinction-to-backscatter ratio of tropospheric aerosol: A numerical study, J. Atmos. Ocean. Technol., 15, 1043-1050.
- Anderson, T. L., Masonis, S. J., Covert, D. S., Charlson, R. J. and Rood, M. J., 2000: In-situ measurement of the aerosol extinction-to-backscatter ratio at a polluted, continental site, J. Geophys. Res., submitted, April, 2000.
- Ansmann, A., Riebesell, M., Wandinger, U., Weitkamp, C., Voss, E., Lahmann, W. and Michaelis, W., 1992: Combined Raman elastic-backscatter LIDAR for vertical profiling of moisture, aerosol extinction, backscatter, and LIDAR ratio, *Appl. Phys. B*, **55**, 18-28.
- Doherty, S., Anderson, T. L. and Charlson, R. J., 1999: Measurement of the lidar ratio for atmospheric aerosols using a 180°-backscatter nephelometer, *Appl. Optics*, **38**, 1823-1832.
- Ferrare, R. A., Melfi, S. H., Whiteman, D. N., Evans, K. D. and Leifer, R., 1998: Raman lidar measurements of aerosol extinction and backscattering 1. Methods comparison, *J. Geophys. Res.*, 103, 19663-19672.
- Kent, G. S., Trepte, C. R., Skeens, K. M. and Winker, D. M., 1998: LITE and SAGE II measurements of aerosols in the southern hemisphere upper troposphere, *J. Geophys. Res.*, 103, 19111-19127.

- Muller, D., Wandinger, U., Althausen, D., Mattis, I. and Ansmann, A., 1998: Retrieval of physical properties from lidar observations of extinction and backscatter at multiple wavelengths, *Appl. Optics*, 37, 2260-2263.
- NRC (1996). <u>Aerosol Radiative Forcing and Climate Change</u>. Washington D. C., National Research Council, National Academy Press.
- Reagan, J. A., Apte, M. V., Bruhns, T. V. and Youngbluth, O., 1984: Lidar and balloon-borne cascade impactor measurements of aerosols: A case study, *Aerosol Sci. Technol.*, 3, 259-275.
- Reagan, J. A., Apte, M. V., Ben-David, A. and Herman, B. M., 1988: Assessment of aerosol extinction to backscatter ratio measurements made at 694.3 nm in Tucson, Arizona, *Aerosol Sci. Technol.*, 8, 215-226.
- Rosen, J. M. and Kjome, T., 1997b: Balloon-borne measurements of the aerosol extinction-to-backscatter ratio, J. Geophys. Res., 102, 11165-11169.
- Rosen, J. M., Pinnick, R. G. and Garvey, D. M., 1997a: Measurement of extinction-to-backscatter ratio for near-surface aerosols, *J. Geophys. Res.*, **102**, 6017-6024.
- Sasano, Y. and Browell, E. V., 1989: Light scattering characteristics of various aerosol types derived from multiple wavelength lidar observations, *Appl. Optics*, **28**, 1670-1679.
- Spinhirne, J. D., Reagan, J. A. and Herman, B. M., 1980: Vertical distribution of aerosol extinction cross section and inference of aerosol imaginary index in the troposphere by lidar technique, *J. Appl. Meteo.*, 19, 426-438.
- Winker, D. M., Couch, R. H. and McCormick, P., 1996: An overview of LITE: NASA's Lidar In-space Technology Experiment, *Proc. IEEE*, **84**, 164-180.
- Young, S. A., Cutten, D. R., Lynch, M. J. and Davies, J. E., 1993: Lidar-derived variations in the backscatter-to-extinction ratio in Southern Hemisphere coastal maritime aerosols, *Atmos. Environ.*, 27A, 1541-1551.

Tables

Table 1: Measurements of S for tropospheric aerosols

	Location	N ^(a)	S (sr)
Method			
slant-path lidar (b)	mixed layer, west U.S.	81	8-75
	(Tucson, AZ)		
horizontal lidar (c)	Netherlands	10	10-50
horizontal lidar (d)	marine surface layer,	10	40-80
	Australian coast		
multi-wavelength	marine boundary layer	1	<30
lidar (e)	(tropical Atlantic)		
` '	mixed layer over	1	43-60
	rainforest (S. Amer.)		
	Saharan dust aloft	2	15-62
	(tropical Atlantic)		
backscatter-sonde (f)	rural, arid SW U.S.	12	42
	rural western U.S.	4	15-30
	lower troposphere		
	rural western U.S.	4	15-60
	upper troposphere		
space-borne lidar (g)	smoke layers in SH	7	50-90
	upper troposphere		
Raman lidar (h)	polluted lower tropo.	2	20-40
	over Northern Germany		
Raman lidar (i)	polluted boundary layer	1	55-95
	over Leipzig, Germany		
Raman lidar (j)	rural, lower tropo. over	25	20-80
•	Oklahoma		
this work (k)	Northwest Coastal U.S.	3	20-70
	Indian Ocean	50	40-90
this work (l)			
this work (I) this work (m)	polluted, Central U.S.	70	30-71

- a Approximate number of independent samples.
- b Determines effective S over mixed layer at 694 nm. (Spinhirne et al., 1980; Reagan et al., 1984; 1988)
- c 1064 nm laser. No information on location of instrument or type of aerosol investigated. (de Leeuw et al., 1986)
- d 532 nm laser beam aimed 2 m over ocean surface from coastal site. Effect of waves not assessed. (Young et al., 1993).
- e S at 600 nm is constrained by Mie calculations based on the measured wavelength variation of β_p . (Sasano and Browell, 1989)
- f S reported at 690 nm. Method requires significant wavelength and angular adjustments, based on assumed size distributions and Mie calculations. (Rosen et al., 1997a, b)
- g S at 532 nm is constrained to values yielding physically plausible lidar retrievals under the assumption of constant S throughout upper troposphere. (Kent et al., 1998)
- h S at 308 nm determined from independent extinction measurements using nitrogen-Raman; one profile. (Ansmann et al., 1992)
- i S at 532 nm using nitrogen-Raman as above. (Muller et al., 1998)
- j S at 351 nm using nitrogen-Raman as above; profiles in lower troposphere over 8 nights (Ferrare et al., 1998)
- k S at 532 nm using the nephelometric technique discussed herein. S was ca. 20 during marine flow and 60-70 during continental flow. (Doherty et al., 1999)
- 1 As in k except for airborne measurements during the Indian Ocean Experiment.
- m As in k except for surface measurements in Central Illinois.
- n As in k except for surface measurements on the windward coast of Oahu.

Figures

Figure 1. Laboratory calibration of in-situ measurement

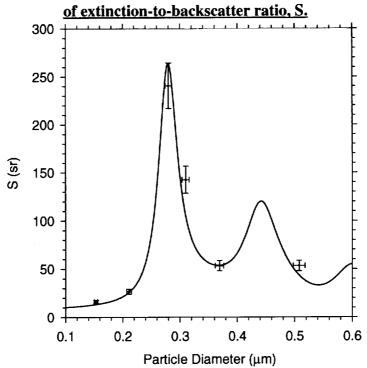
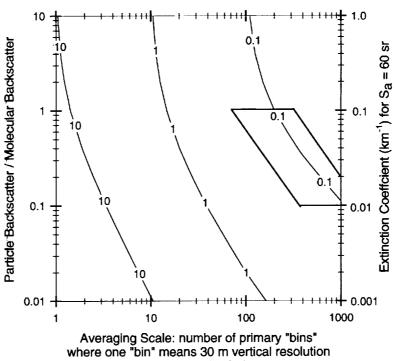


Figure 2: Relative Uncertainty in 532 nm Particle Backscatter



Averaging Scale: number of primary "bins" where one "bin" means 30 m vertical resolution and a single shot